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## STATEMENT

I, Yasuhide KOBAYASHI, residing at 2-24-12, Hachimanyama, Setagaya-ku, Tokyo, Japan, hereby state that I have a thorough knowledge of the English and Japanese languages and that the attached document is an accurate English translation of the Japanese specification of Japanese Patent Application No. Hei.-11-370349 filed December 27, 1999, upon which the present application claims a priority.

Declared at Tokyo, Japan

This 14th day of August, 2003

A handwritten signature in black ink, appearing to read "Y. Kobayashi".

Yasuhide Kobayashi

**PATENT OFFICE  
JAPANESE GOVERNMENT**

This is to certify that the annexed is a true copy of the following application as filed with this Office.

Date of Application: December 27, 1999

Application Number: Japanese Patent Application  
No. 11-370349

Applicant(s): FUJI PHOTO FILM CO., LTD.

April 13, 2001  
Commissioner,  
Patent Office: Kozo OIKAWA (Seal)

Issuance No. 2001-3029556

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[Date of Filing]	December 27, 1999
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[International Classification]	H05B 33/14
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[List of Documents]

[Name of Document]	Specification	1 copy
[Name of Document]	Abstract	1 copy
[General Power of Attorney No.]	9723355	

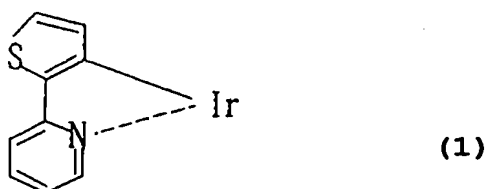
[Designation of Document]      Specification

[Title of the Invention]

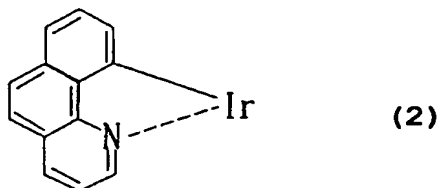
LIGHT-EMITTING MATERIAL COMPRISING ORTHOMETALATED IRIDIUM  
COMPLEX, AND LIGHT-EMITTING ELEMENT

[Claims]

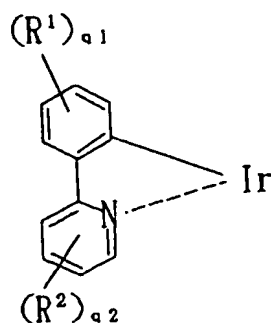
1. A light-emitting material comprising a compound having a partial structure represented by the following general formula (1) or tautomer thereof:



2. A light-emitting material comprising a compound having a partial structure represented by the following general formula (2) or tautomer thereof:



3. A light-emitting material comprising a compound having a partial structure represented by the following general formula (3) or tautomer thereof:



(3)

wherein  $R^1$  and  $R^2$  each represent a substituent; and  $q^1$  and  $q^2$  each represent an integer of from 0 to 4, with the proviso that the sum of  $q^1$  and  $q^2$  is 1 or more.

4. An organic light-emitting element comprising a light-emitting layer or a plurality of thin organic compound layers containing a light-emitting layer formed interposed between a pair of electrodes, characterized in that at least one of said thin organic compound layers contains a light-emitting material according to any one of Claims 1, 2 and 3.

5. An organic light-emitting element comprising a light-emitting layer or a plurality of thin organic compound layers containing a light-emitting layer formed interposed between a pair of electrodes, characterized in that at least one of said thin organic compound layers contains an orthometalated iridium complex and said layer containing an orthometalated iridium complex is formed by a coating process.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The present invention relates to a light-emitting

material and light-emitting element capable of converting electric energy to light which is then emitted and more particularly to a light-emitting element which can be preferably used in various arts such as display element, display, backlight, electrophotography, illuminating light source, recording light source, exposure light source, reading light source, sign, advertising display and interior.

[0002]

[Prior Art]

Today, various display elements have been under active study and development. In particular, an organic electric field light-emitting (EL) element can emit with a high luminance at a low voltage and thus has been noted as a favorable display element. For example, a light-emitting element having a vacuum-deposited thin organic layer has been known (Applied Physics Letters, vol. 51, page 913, 1987). The light-emitting element described in this reference comprises as an electron-transporting material tris(8-hydroxyquinolate) aluminum complex (Alq) which is laminated with a positive hole-transporting material (amine compound) to exhibit drastically improved light-emitting properties as compared with the conventional single-layer type elements.

[0003]

In recent years, the application of organic EL element to color display has been under active study. However, in order



to develop a high performance color display, it is necessary that the properties of blue, green and red light-emitting elements be each improved.

[0004]

As a means for improving the properties of light-emitting elements there has been reported a green light-emitting element utilizing the emission of light from orthometalated iridium complex ( $\text{Ir(ppy)}_3$ : Tris-Ortho-Metalated Complex of Iridium (III) with 2-Phenylpyridine) (Applied Physics Letters 75, 4 (1999)). The foregoing element can attain an external quantum yield of 8%, which is higher than the limit of the external quantum yield of the conventional light-emitting elements, i.e., 5%. However, since the foregoing light-emitting element is limited to green light-emitting element, the range within it can be applied as a display is narrow. It has thus been desired to develop light-emitting materials capable of emitting light having other colors.

[0005]

On the other hand, an organic light-emitting element which can attain light emission with a high luminance is one having a laminate of vacuum-deposited organic material layers. The preparation of such an element is preferably accomplished by a coating method from the standpoint of simplification of production procedure, workability, area attained, etc. However, the element prepared by the conventional coating method

is inferior to that prepared by vacuum evaporation method particularly in light-emitting efficiency. It has thus been desired to develop a novel light-emitting material.

[0006]

[Problems that the Invention is to Solve]

An object of the present invention is to provide a light-emitting element having good light-emitting properties, a light-emitting material which can form such a light-emitting element.

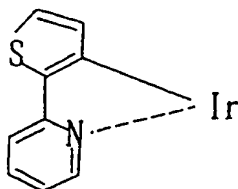
[0007]

[Means for Solving the Problems]

The foregoing object of the invention can be accomplished by the following means.

1. A light-emitting material comprising a compound having a partial structure represented by the following general formula (1) or tautomer thereof:

[0008]

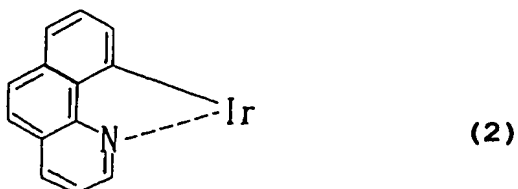


(1)

[0009]

2. A light-emitting material comprising a compound having a partial structure represented by the following general formula (2) or tautomer thereof:

[0010]

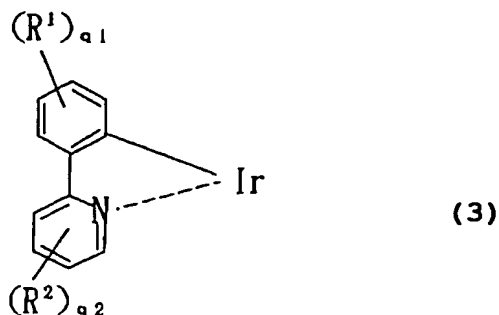


[0011]

3. A light-emitting material comprising a compound having a partial structure represented by the following general formula (3) or tautomer thereof:

wherein  $R^1$  and  $R^2$  each represent a substituent; and  $q^1$  and  $q^2$  each represent an integer of from 0 to 4, with the proviso that the sum of  $q^1$  and  $q^2$  is 1 or more.

[0012]



[0013]

4. An organic light-emitting element comprising a light-emitting layer or a plurality of thin organic compound layers containing a light-emitting layer formed interposed

between a pair of electrodes, characterized in that at least one of the thin organic compound layers contains a light-emitting material according to any one of Clauses 1, 2 and 3.

5. An organic light-emitting element comprising a light-emitting layer or a plurality of thin organic compound layers containing a light-emitting layer formed interposed between a pair of electrodes, characterized in that at least one of the thin organic compound layers contains an orthometalated iridium complex and said layer containing an orthometalated iridium complex is formed by a coating process.

[0014]

[Mode for Carrying Out the Invention]

The present invention will be further described hereinafter.

The compound according to the invention is a light-emitting material comprising an orthometalated iridium complex. "Orthometalated metal complex" is a generic term for a group of compounds as described in Akio Yamamoto, "Yuki Kinzoku Kagaku- Kiso to Oyo- (Organic Metal Chemistry - Fundamentals and Application)", Shokabosha, pp. 150, 232, 1982, H. Yersin, "Photochemistry and Photophysics of Coordination Compounds", Springer-Verlag, pp. 1 - 77, pp. 135 - 146, 1987, etc.

[0015]

The valence of iridium in the orthometalated iridium complex is not specifically limited but is preferably 3. The

ligands constituting the orthometalated iridium complex are not specifically limited so far as they can form an orthometalated complex. In practice, however, there may be used, e.g., aryl group-substituted nitrogen-containing heterocyclic derivative (The aryl group substitutes for the nitrogen-containing heterocycle on the carbon atom adjacent to nitrogen atom. Examples of the aryl group include phenyl group, naphthyl group, anthracenyl group, and pyrenyl group. Examples of the nitrogen-containing heterocycle include pyridine, pyrimidine, pyrazine, pyridazine, quinoline, isoquinoline, quinoxaline, phthalazine, quinazoline, naphtholidine, cinnoline, perimidine, phenanthroline, pyrrole, imidazole, pyrazole, oxazole, oxadiazole, triazole, thiadiazole, benzimidazole, benzoxazole, and phenanthridine), heteroaryl group-substituted nitrogen-containing heterocyclic derivative (The heteroaryl group substitutes for the nitrogen-containing heterocycle on the carbon atom adjacent to nitrogen atom. Examples of the heteroaryl group include group containing the foregoing nitrogen-containing heterocyclic derivative, chenyl group, and furyl group), 7,8-benzoquinoline derivative, phosphinoaryl derivative, phosphinoheteroaryl derivative, phosphinoxyaryl derivative, phosphinoxyheteroaryl derivative, aminomethylaryl derivative, aminomethylheteroaryl derivative, etc. Preferred among these ligands are aryl group-substituted nitrogen-containing

aromatic heterocyclic derivative, heteroaryl group-substituted nitrogen-containing aromatic heterocyclic derivative, and 7,8-benzoquinoline derivative. Even more desirable among these ligands are phenylpyridine derivative, thiophenylpyridine derivative, and 7,8-benzoquinoline derivative. Particularly preferred among these ligands are thiophenylpyridine derivative, and 7,8-benzoquinoline derivative.

[0016]

The compound of the invention may have ligands other than the ligands required to form an orthometalated complex. Examples of the other ligands include various known ligands. Examples of these ligands include those described in H. Yersin, "Photochemistry and Photophysics of Coordination Compounds", Springer-Verlag, 1987, Akio Yamamoto, "Yuki Kinzoku Kagaku-Kiso to Oyo- (Organic Metal Chemistry - Fundamentals and Application)", Shokabosha, 1982, etc. Preferred among these ligands are halogen ligands (preferably chlorine ligand), nitrogen-containing heterocyclic ligands (e.g., bipyridyl, phenanthroline), and diketone ligands. Even more desirable among these ligands are chlorine ligand and bipyridyl ligand.

[0017]

There may be used one or a plurality of kinds of ligands constituting the compound of the invention. The number of ligands in the complex is preferably from 1 to 3, particularly

from 1 or 2, more preferably 1.

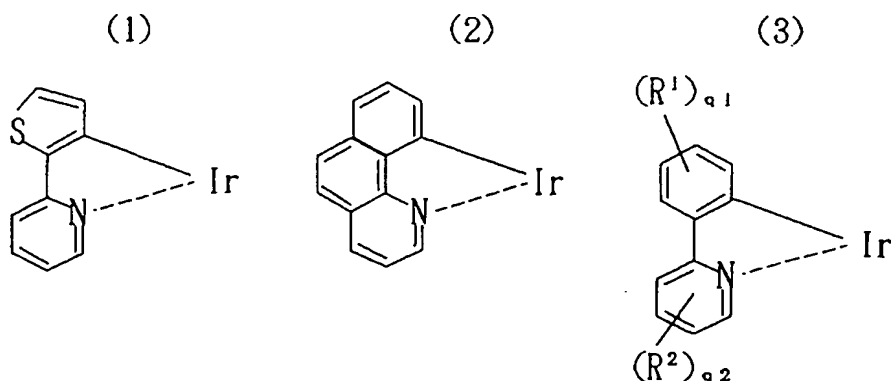
[0018]

The number of carbon atoms in the compound of the invention is preferably from 5 to 100, more preferably from 10 to 80, even more preferably from 14 to 50.

[0019]

Preferred embodiment of the compounds of the invention is: a compound having a partial structure represented by the general formula (1) or tautomer thereof; a compound having a partial structure represented by the general formula (2) or tautomer thereof; or a compound having a partial structure represented by the general formula (3) or tautomer thereof. The compound having a partial structure represented by the general formula (1) or tautomer thereof; and the compound having a partial structure represented by the general formula (2) or tautomer thereof are preferred.

[0020]



[0021]

The compound having a partial structure represented by the general formula (1) or tautomer thereof may have one iridium atom per molecule or may be a so-called binuclear complex having two or more iridium atoms per molecule. This compound may further contain other metal atoms. This can apply to the compounds having a partial structure represented by the general formula (2) to (3) or tautomers thereof.

[0022]

In the general formula (3),  $R^1$  and  $R^2$  each represents a substituent. The suffixes  $q^1$  and  $q^2$  each represent an integer of from 0 to 4, with the proviso that the sum of  $q^1$  and  $q^2$  is 1 or more. When  $q^1$  and  $q^2$  each are 2 or more, the plurality of  $R^1$ 's and  $R^2$ 's may be the same or different.

[0023]

Examples of the group represented by  $R^1$  or  $R^2$  include alkyl group (alkyl group preferably having from 1 to 30, more preferably from 1 to 20, particularly from 1 to 10 carbon atoms, e.g., methyl, ethyl, iso-propyl, tert-butyl, n-octyl, n-decyl, n-hexadecyl, cyclopropyl, cyclopentyl, cyclohexyl), alkenyl group (alkenyl group preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, particularly from 2 to 10 carbon atoms, e.g., vinyl, allyl, 2-butenyl, 3-pentenyl), alkynyl group (alkynyl group preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms,



particularly from 2 to 10 carbon atoms, e.g., propargyl, 3-pentynyl), aryl group (aryl group preferably having from 6 to 30 carbon atoms, more preferably from 6 to 20 carbon atoms, particularly from 6 to 12 carbon atoms, e.g., phenyl, p-methylphenyl, naphthyl, anthranyl), amino group (amino group preferably having from 0 to 30 carbon atoms, more preferably from 0 to 20 carbon atoms, particularly from 0 to 10 carbon atoms, e.g., amino, methylamino, dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino), alkoxy group (alkoxy group preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, particularly from 1 to 10 carbon atoms, e.g., methoxy, ethoxy, butoxy, 2-ethylhexyloxy), aryloxy group (aryloxy group preferably having from 6 to 30 carbon atoms, more preferably from 6 to 20 carbon atoms, particularly from 6 to 12 carbon atoms, e.g., phenyloxy, 1-naphthyloxy, 2-naphthyloxy), heteroaryloxy group (heteroaryloxy group preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., pyridyloxy, pyrazolyloxy, pyrimidyloxy, quinolyloxy), acyl group (acyl group preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., acetyl, benzoyl, formyl, pivaloyl), alkoxycarbonyl group (alkoxycarbonyl group preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, particularly from 2 to 12 carbon

atoms, e.g., methoxycarbonyl, ethoxycarbonyl), aryloxy carbonyl group (aryloxy carbonyl group preferably having from 7 to 30 carbon atoms, more preferably from 7 to 20 carbon atoms, particularly from 7 to 12 carbon atoms, e.g., phenyloxy carbonyl), acyloxy group (acyloxy group preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, particularly from 2 to 10 carbon atoms, e.g., acetoxy, benzoyloxy), acylamino group (acylamino group preferably having from 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly from 2 to 10 carbon atoms, e.g., acetylamino, benzoylamino), alkoxycarbonylamino group (alkoxycarbonylamino group preferably having from 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly from 2 to 12 carbon atoms, e.g., methoxycarbonylamino), aryloxy carbonylamino group (aryloxy carbonylamino group preferably having from 7 to 30 carbon atoms, more preferably 7 to 20 carbon atoms, particularly from 7 to 12 carbon atoms, e.g., phenyloxy carbonylamino), sulfonylamino group (sulfonylamino group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., methanesulfonylamino, benzenesulfonylamino), sulfamoyl group (sulfamoyl group preferably having from 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, particularly from 2 to 10 carbon atoms, e.g., sulfamoyl, methylsulfamoyl, dimethylsulfamoyl,

phenylsulfamoyl), carbamoyl group (carbamoyl group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., carbamoyl, methylcarbamoyl, diethylcarbamoyl, phenylcarbamoyl), alkylthio group (alkylthio group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., methylthio, ethylthio), arylthio group (arylthio group preferably having from 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, particularly from 6 to 12 carbon atoms, e.g., phenylthio), heteroarylthio group (heteroarylthio group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., pyridylthio, 2-benzimidazolylthio, 2-benzoxazolylthio, 2-benzthiazolylthio), sulfonyl group (sulfonyl group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., mesyl, tosyl), sulfinyl group (sulfinyl group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., methanesulfinyl, benzenesulfinyl), ureide group (ureide group preferably having from 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., ureide, methylureide, phenylureide), phosphoric acid amide group (phosphoric acid amide group preferably having from 1 to 30 carbon atoms, more preferably

1 to 20 carbon atoms, particularly from 1 to 12 carbon atoms, e.g., diethylphosphoric acid amide, phenylphosphoric acid amide), hydroxy group, mercapto group, halogen atom (e.g., fluorine atom, chlorine atom, bromine atom, iodine atom), cyano group, sulfo group, carboxyl group, nitro group, hydroxamic acid group, sulfinio group, hydrazino group, imino group, heterocyclic group (heterocyclic group preferably having from 1 to 30 carbon atoms, more preferably from 1 to 12 carbon atoms, and containing as hetero atoms nitrogen atom, oxygen atom and sulfur atom, e.g., imidazolyl, pyridyl, quinolyl, furyl, chenyl, piperidyl, morpholino, benzoxazolyl, benzimidazolyl, benzthiazolyl), and silyl group (silyl group preferably having from 3 to 40 carbon atoms, more preferably 3 to 30 carbon atoms, particularly from 3 to 24 carbon atoms, e.g., trimethylsilyl, triphenylsilyl). These substituents may be further substituted.  $R^1$ 's or  $R^2$ 's may be connected to each other, or  $R^1$  and  $R^2$  may be connected to each other to form a condensed ring structure.

[0024]

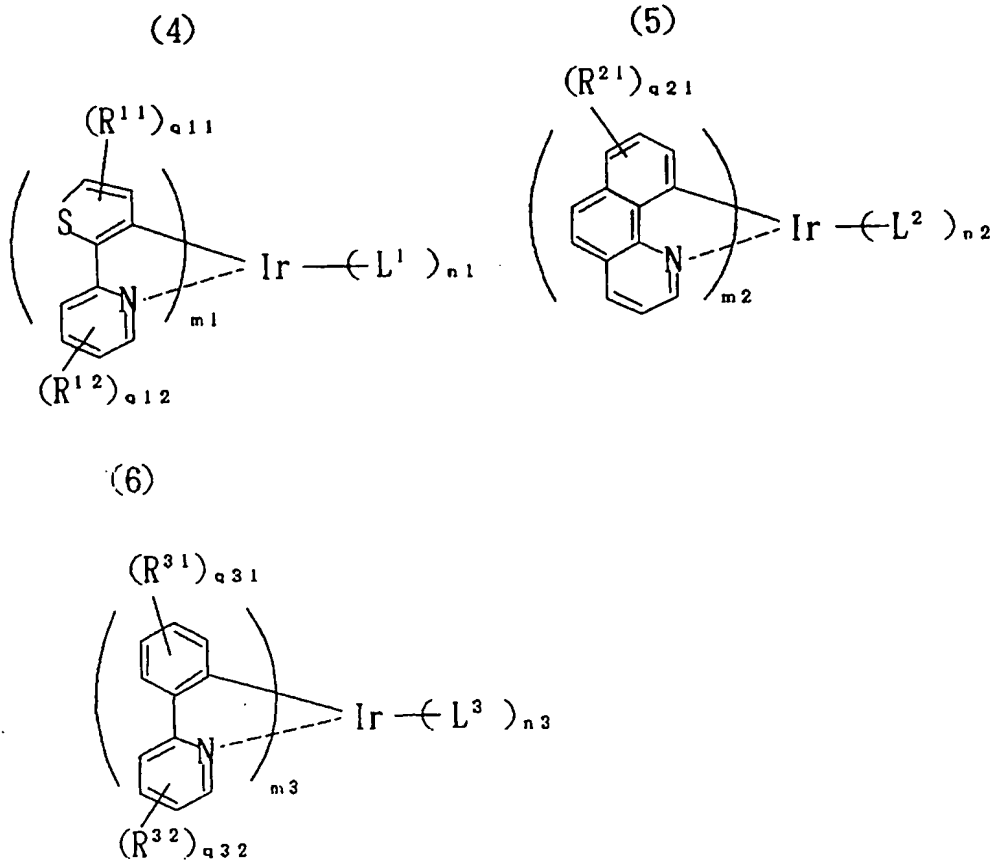
$R^1$  and  $R^2$  each are preferably an alkyl group, aryl group, alkoxy group, or a group which forms a condensed ring structure when  $R^1$  and  $R^2$  are connected to each other. Preferred among these groups are alkyl group and group which forms a condensed ring structure when  $R^1$  and  $R^2$  are connected to each other. The suffixes  $q^1$  and  $q^2$  each are preferably 0, 1 or 2. More preferably,

the sum of  $q^1$  and  $q^2$  is 1 or 2.

[0025]

Further preferred embodiment of the compounds of the invention is a compound represented by the general formula (4), a compound represented by the general formula (5) and a compound represented by the general formula (6). The compound represented by the general formula (4) and the compound represented by the general formula (5) are particularly preferred.

[0026]



[0027]

The general formula (4) will be further described hereinafter.  $R^{11}$  and  $R^{12}$  each represent a substituent. Examples of the substituent represented by  $R^{11}$  or  $R^{12}$  include those described with reference to  $R^1$  above.

[0028]

$R^{11}$  and  $R^{12}$  each are preferably an alkyl or aryl group, more preferably an alkyl group.

[0029]

The suffix  $q^{11}$  represents an integer of from 0 to 2, preferably 0 or 1, more preferably 0. The suffix  $q^{12}$  represents an integer of from 0 to 4, preferably 0 or 1, more preferably 0. When  $q^{11}$  and  $q^{12}$  each are 2 or more, the plurality of  $R^{11}$ 's and  $R^{12}$ 's may be the same or different or may be connected to each other to form a condensed ring.

[0030]

$L^1$  represents a ligand. Examples of such a ligand include ligands required to form the foregoing orthometalated iridium complexes and ligands described with reference to other ligands.  $L^1$  is preferably a ligand required to form an orthometalated iridium complex, nitrogen-containing heterocyclic ligand, diketone ligand or halogen ligand, more preferably ligand required to form an orthometalated iridium complex or bipyridyl ligand.

[0031]

The suffix  $n^1$  represents an integer of from 0 to 5, preferably 0. The suffix  $m^1$  represents an integer of from 1 to 3, preferably 3. The combination of  $n^1$  and  $m^1$  is preferably such that the metal complex represented by the general formula (4) is a neutral complex.

[0032]

The general formula (5) will be further described hereinafter.  $R^{21}$ ,  $n^2$ ,  $m^2$ , and  $L^2$  have the same meaning as  $R^{11}$ ,  $n^1$ ,  $m^1$ , and  $L^1$ , respectively. The suffix  $q^{21}$  represents an integer of from 0 to 8, preferably 0. When  $q^{21}$  is 2 or more, the plurality of  $R^{21}$ 's may be the same or different or may be connected to each other to form a condensed ring.

[0033]

The general formula (6) will be further described hereinafter.  $R^{31}$ ,  $R^{32}$ ,  $q^{31}$ ,  $q^{32}$ ,  $n^3$ ,  $m^3$ , and  $L^3$  have the same meaning as  $R^1$ ,  $R^2$ ,  $q^1$ ,  $q^2$ ,  $n^1$ ,  $m^1$ , and  $L^1$ , respectively.

[0034]

The compound of the invention may be a so-called low molecular compound having one repeating unit such as one represented by the general formula (1) or may be a so-called oligomer or polymer compound having a plurality of repeating units such as one represented by the general formula (1) (having a weight-average molecular weight (in polystyrene equivalence) of preferably from 1,000 to 5,000,000, more preferably from 2,000 to 1,000,000, even more preferably from 3,000 to 100,000).

The compound of the invention is preferably a low molecular compound.

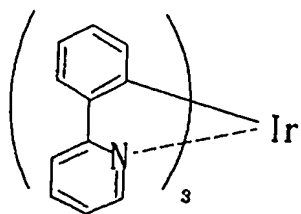
[0035]

Examples of the compound to be used in the invention will be given below, but the present invention should not be construed as being limited thereto.

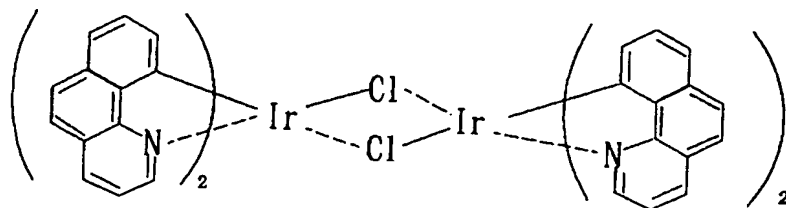


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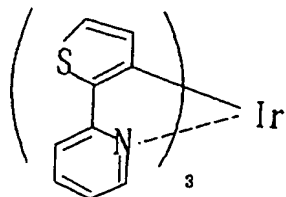
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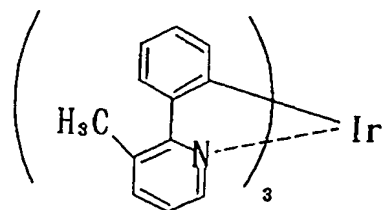
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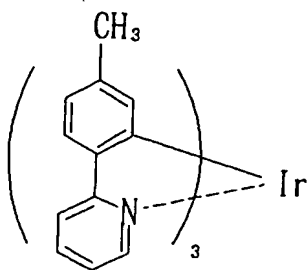
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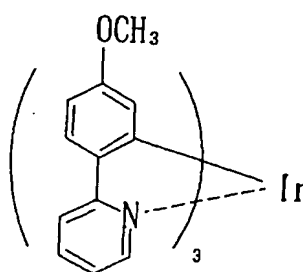
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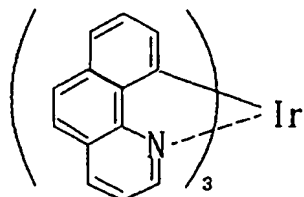


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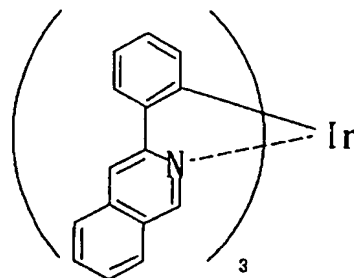


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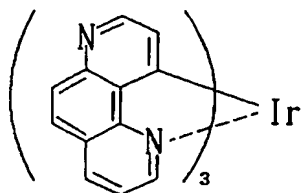
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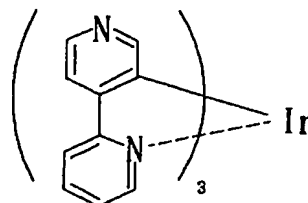
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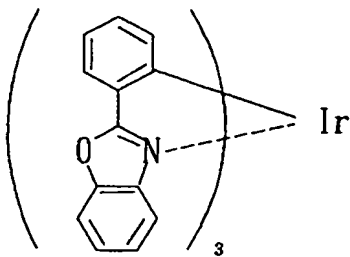
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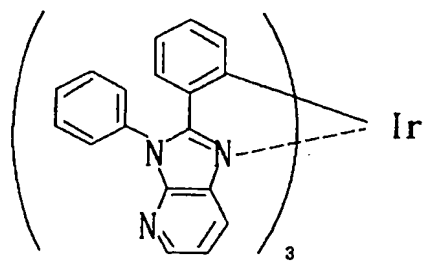
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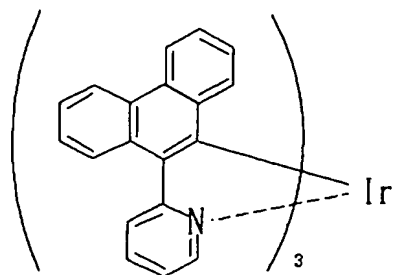
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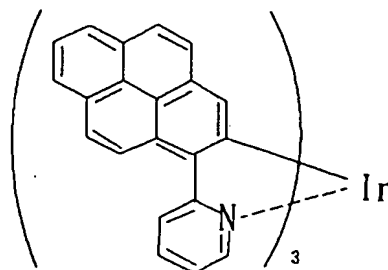
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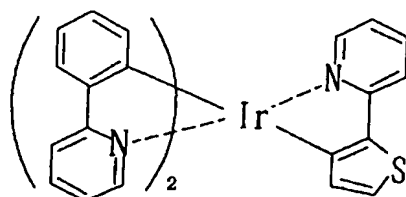


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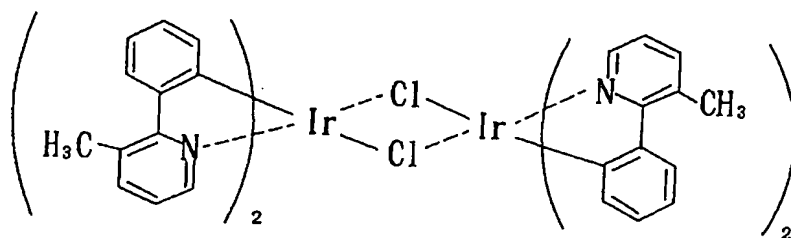


[0038]

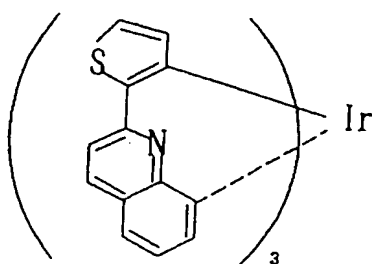
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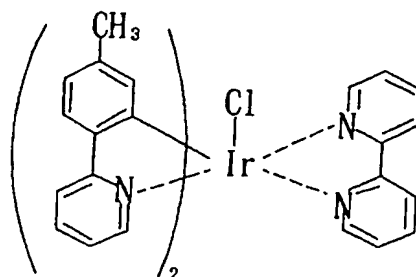
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(1-17)

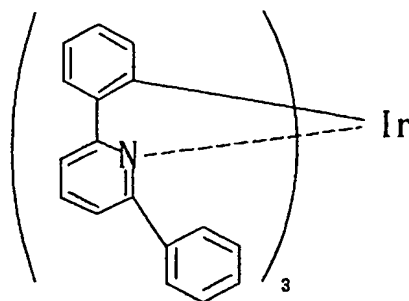


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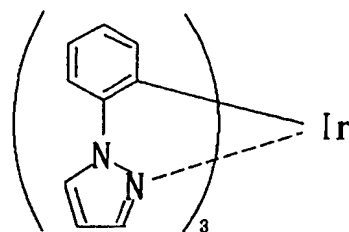


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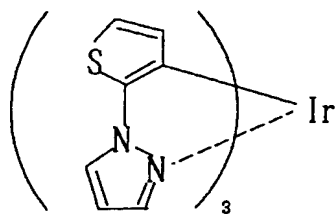
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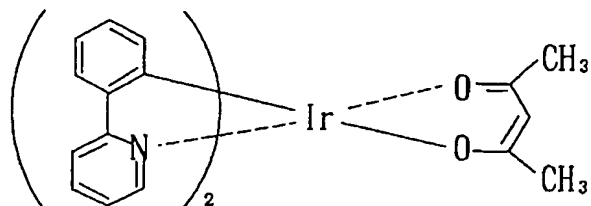
(1-20)



(1-21)



(1-22)



[0040]

The synthesis of the compound of the invention can be accomplished by any known method as disclosed in "Inorg. Chem.", No. 30, page 1,685, 1991, No. 27, page 3,464, 1988, No. 33, page 545, 1994, "Inorg. Chem. Acta.", No. 181, page 245, 1991, "J. Organomet. Chem.", No. 35, page 293, 1987, "J. Am. Chem. Soc.", No. 107, page 1,431, 1985, etc.

[0041]

The light-emitting element comprising the compound of the invention will be further described hereinafter. The light-emitting element of the invention is not specifically limited in its system, driving method and form of utilization so far as it comprises the compound of the invention. In practice, however, the light-emitting element of the invention is preferably in the form of structure utilizing light emission from the compound of the invention or structure comprising the compound of the invention as a charge-transporting material. A representative example of light-emitting element is an organic EL (electroluminescence) element.

[0042]

The process for the formation of the organic layer in the light-emitting element comprising the compound of the invention is not specifically limited. In practice, however, any method such as resistively-heated vacuum evaporation method, electron beam method, sputtering method, molecular lamination

method, coating method, ink jet method and printing method may be used. Preferred among these methods are resistively-heated vacuum evaporation method and coating method from the standpoint of properties and producibility. More desirable among these methods is coating method from the standpoint of prevention of thermal decomposition during vacuum evaporation.

[0043]

The light-emitting element of the invention comprises a light-emitting layer or a plurality of thin organic compound layers containing a light-emitting layer formed interposed between a pair of electrodes, i.e., cathode and anode. There may be provided a positive hole-injecting layer, a positive hole-transporting layer, an electron-injecting layer, an electron-transporting layer and a protective layer besides the light-emitting layer. These layers may be provided with other functions. The various layers may be each made of various materials.

[0044]

The anode supplies a positive hole into the positive hole-injecting layer, positive hole-transporting layer, light-emitting layer, etc. The anode may be made of a metal, alloy, metal oxide, electrically-conductive compound or mixture thereof, preferably a material having a work function of 4 eV or more. Specific examples of such a material include electrically-conductive metal oxide such as tin oxide, zinc

oxide, indium oxide and indium tin oxide (ITO), metal such as gold, silver, chromium and nickel, mixture or laminate of such a metal and electrically-conductive metal oxide, electrically inorganic material such as copper iodide and copper sulfate, electrically-conductive organic material such as polyaniline, polythiophene and polypyrrole, and laminate of these materials with ITO. Preferred among these materials are electrically-conductive metal oxides. Particularly preferred among these electrically-conductive metal oxides is ITO from the standpoint of producibility, electrical conductivity and transparency. The thickness of the anode may be properly predetermined depending on its material. In practice, however, it is preferably from 10 nm to 5  $\mu$ m, more preferably from 50 nm to 1  $\mu$ m, even more preferably from 100 nm to 500 nm.

[0045]

The anode is normally used in the form of anode layer formed on soda-lime glass, non-alkali glass, transparent resin substrate or the like. As the glass, if used, there is preferably used non-alkali glass to reduce the amount of ions to be eluted therefrom. Soda-lime glass, if used, is preferably coated with a barrier such as silica. The thickness of the substrate is not specifically limited so far as it suffices to maintain a desired mechanical strength. In practice, however, it is normally 0.2 mm or more, preferably 0.7 mm if glass is used.

The preparation of the anode may be accomplished by any method depending on the materials used. If ITO is used, for example, electron beam method, sputtering method, resistively-heated vacuum evaporation method, chemical reaction method (sol-gel method), method involving the coating of a dispersion of indium tin oxide or the like can be used to form an anode layer.

The anode can be cleaned or otherwise treated to lower the driving voltage of the element or enhance the light emission efficiency of the element. The anode made of ITO, for example, can be effectively subjected to UV-ozone treatment, plasma treatment, etc.

[0046]

The cathode supplies electron into the electron-injecting layer, electron-transporting layer, light-emitting layer, etc. The cathode is selected taking into account the adhesivity to the layer adjacent to the negative electrode such as electron-injecting layer, electron-transporting layer and light-emitting layer, ionization potential, stability, etc. As the material constituting the cathode there may be used a metal, alloy, metal halide, metal oxide, electrically-conductive compound or mixture thereof. Specific examples of such a material include alkaline metal (e.g., Li, Na, K), fluoride thereof, alkaline earth metal (e.g., Mg, Ca), fluoride thereof, gold, silver,



lead, aluminum, sodium-potassium alloy, mixture thereof, lithium-aluminum alloy, mixture thereof, magnesium-silver alloy, mixture thereof, and rare earth metal such as indium and ytterbium. Preferred among these materials are those having a work function of 4 eV or less. Even more desirable among these materials are aluminum, lithium-aluminum alloy, mixture thereof, magnesium-silver alloy, and mixture thereof. The cathode may be not only in the form of single layer structure comprising the foregoing compound or mixture but also in the form of laminated structure comprising the foregoing compound or mixture. The thickness of the cathode may be properly predetermined depending on its material. In practice, however, it is preferably from 10 nm to 5  $\mu\text{m}$ , more preferably from 50 nm to 1  $\mu\text{m}$ , even more preferably from 100 nm to 1  $\mu\text{m}$ .

The preparation of the cathode can be accomplished by any method as electron beam method, sputtering method, resistively-heated vacuum evaporation method and coating method. A single metal may be vacuum-vaporized. Alternatively, two or more components may be vacuum-vaporized at the same time. Further, a plurality of metals may be vacuum-vaporized to form an alloy electrode. Alternatively, an alloy which has been previously prepared may be vacuum-vaporized.

The sheet resistivity of the anode and cathode is preferably as low as possible and thus is preferably hundreds

of ohm/□ or less.

[0047]

As the material constituting the light-emitting layer there may be used any material which can form a layer capable of injecting positive hole from the anode, positive hole-injecting layer or positive hole-transporting layer as well as injecting electron from the cathode, electron-injecting layer or electron-transporting layer during the application of electric field, moving electron thus injected or providing a site for the recombination of positive hole and electron for emission of light. Alternatively, any material which emits light from either singlet exciton or triplet exciton may be used. Examples of the light-emitting material employable herein include various metal complexes such as metal complex and rare earth complex of benzoxazole derivative, benzoimidazole derivative, benzothiazole derivative, styrylbenzene derivative, polyphenyl derivative, diphenylbutadiene derivative, tetraphenylbutadiene derivative, naphthalimide derivative, coumarine derivative, perylene derivative, perynone derivative, oxadiazole derivative, aldazine derivative, pyralidine derivative, cyclopentadiene derivative, bisstyrylanthracene derivative, quinacridone derivative, pyrrolopyridine derivative, thiadiazolopyridine derivative, cyclopentadiene derivative, styrylamine derivative, aromatic dimethylidine compound and

8-quinolinol derivative, polymer compound such as polythiophene, polyphenylene and polyphenylenevinylene, organic silane derivative, and the compound of the invention. The thickness of the light-emitting layer is not specifically limited but is normally from 1 nm to 5  $\mu\text{m}$ , preferably from 5 nm to 1  $\mu\text{m}$ , even more preferably from 10 nm to 500 nm.

The process for the formation of the light-emitting layer is not specifically limited. In practice, however, any method such as resistively-heated vacuum evaporation method, electron beam method, sputtering method, molecular lamination method, coating method (e.g, spin coating method, casting method, dip coating method), ink jet method, LB method and printing method may be used. Preferred among these methods are resistively-heated vacuum evaporation method and coating method.

[0048]

As the material constituting the positive hole-injecting layer and positive hole-transporting layer there may be used any material having any of capability of injecting positive hole from the anode, capability of transporting positive hole and capability of giving barrier to electron injected from the cathode. Specific examples of such a material include electrically-conductive polymer oligomers such as carbazole derivative, triazole derivative, oxazole derivative, oxadiazole derivative, imidazole derivative, polyaryllalkane

derivative, pyrazoline derivative, pyrazolone derivative, phenylenediamine derivative, arylamine derivative, amino-substituted chalcone derivative, styrylanthracene derivative, fluorenone derivative, hydrazone derivative, stilbene derivative, silazalane derivative, aromatic tertiary amine compound, styrylamine compound, aromatic dimethyldine compound, porphyrin compound, polysilane compound, poly(N-vinylcarbazole) derivative, aniline copolymer, thiophene oligomer and polythiophene, organic silane derivative, carbon film, and the compound of the invention. The thickness of the positive hole-injecting layer and positive hole-transporting layer is not specifically limited but is preferably from 1 nm to 5  $\mu\text{m}$ , more preferably from 5 nm to 1  $\mu\text{m}$ , even more preferably from 10 nm to 500 nm. The positive hole-injecting layer and positive hole-transporting layer each may be in the form of single layer structure made of one or more of the foregoing material or multi-layer structure consisting of a plurality of layers having the same or different compositions.

The formation of the positive hole-injecting layer and positive hole-transporting layer can be accomplished by any method such as vacuum evaporation method, LB method, method involving the coating of a solution or dispersion of the foregoing positive hole-injecting or transporting material in a solvent (e.g., spin coating method, casting method, dip coating

method), ink jet method and printing method. In the case of coating method, the foregoing positive hole-injecting or transporting material may be dissolved or dispersed in a solvent with a resin component. Examples of such a resin component include polyvinyl chloride, polycarbonate, polystyrene, polymethyl methacrylate, polybutyl methacrylate, polyester, polysulfone, polyphenylene oxide, polybutadiene, poly(N-vinylcarbazole), hydrocarbon resin, ketone resin, phenoxy resin, polyamide, ethyl cellulose, vinyl acetate, ABS resin, polyurethane, melamine resin, unsaturated polyester resin, alkyd resin, epoxy resin, and silicon resin.

[0049]

As the material constituting the electron-injecting material layer and electron-transporting layer there may be used any material having any of capability of injecting electron from the cathode, capability of transporting electron and capability of giving barrier to positive hole injected from the anode. Specific examples of such a material include various metal complexes such as metal complex of heterocyclic tetracarboxylic anhydride such as triazole derivative, oxazole derivative, oxadiazole derivative, fluorenone derivative, anthraquinodimethane derivative, anthrone derivative, diphenylquinone derivative, thiopyranedioxide derivative, carbodiimide derivative, fluorenilidenemethane derivative, distyrylpyrazine derivative and naphthaleneperylene,

phthalocyanine derivative and 8-quinolinol derivative and metal complex comprising metal phthalocyanine, benzoxazole or benzothiazole as a ligand, and organic silane derivative. The thickness of the electron-injecting layer and electron-transporting layer is not specifically limited but is preferably from 10 nm to 500 nm, more preferably from 5 nm to 1  $\mu\text{m}$ , even more preferably from 10 nm to 500 nm. The electron-injecting layer and electron-transporting layer each may be in the form of single layer structure made of one or more of the foregoing material or multi-layer structure consisting of a plurality of layers having the same or different compositions.

The formation of the electron-injecting layer and electron-transporting layer can be accomplished by any method such as vacuum evaporation method, LB method, method involving the coating of a solution or dispersion of the foregoing positive hole-injecting or transporting material in a solvent (e.g., spin coating method, casting method, dip coating method), ink jet method and printing method. In the case of coating method, the foregoing positive hole-injecting or transporting material may be dissolved or dispersed in a solvent with a resin component.

As the resin component there may be used any of those exemplified with reference to the positive hole-injecting or transporting layer.

[0050]

As the material constituting the protective layer there may be used any material capable of preventing materials which accelerating the deterioration of the element such as water content and oxygen from entering into the element. Specific examples of such a material include metal such as In, Sn, Pb, Au, Cu, Ag, Al, Ti and Ni, metal oxide such as MgO, SiO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, GeO, NiO, CaO, BaO, Fe<sub>2</sub>O<sub>2</sub>, Y<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>, metal fluoride such as MgF<sub>2</sub>, LiF, AlF<sub>3</sub> and CaF<sub>2</sub>, polyethylene, polypropylene, polymethyl methacrylate, polyimide, polyurea, polytetrafluoroethylene, polychlorotrifluoroethylene, polydichlorofluoroethylene, polymer of chlorotrifluoroethylene with dichlorodifluoroethylene, copolymer obtained by the copolymerization of tetrafluoroethylene with a monomer mixture comprising at least one comonomer, fluorine-containing copolymer having a cyclic structure in the copolymer main chain, water-absorbing material having a water absorption of 1% or more, and moisture-resistant material having a water absorption of 0.1% or less.

The process for the formation of the protective layer is not specifically limited. Examples of the method employable herein include vacuum evaporation method, sputtering method, reactive sputtering method, MBE (molecular beam epitaxy) method, cluster ion beam method, ion plating method, plasma polymerization method (high frequency excited ion plating method), plasma CVD method, laser CVD method, heat CVD method,

gas source CVD method, coating method, and printing method.

[0051]

[Example]

Specific embodiments of implication of the invention will be described hereinafter, but the present invention should not be construed as being limited thereto.

[0052]

#### COMPARATIVE EXAMPLE 1

40 mg of a poly(N-vinylcarbazole), 12 mg of PBD (2-(4-biphenyl)-5-(4-t-butylphenyl)-1,3,4-oxadiazole) and 1 mg of the following compound A were dissolved in 2.5 ml of dichloroethane. The solution thus obtained was then spin-coated onto a substrate which had been cleaned (1,500 rpm, 20 sec). The thickness of the organic layer thus formed was 98 nm. A patterned mask (arranged such that the light-emitting area was 4 mm x 5 mm) was then disposed on the thin organic layer. Magnesium and silver were then simultaneously vacuum-evaporated onto the thin organic layer at a ratio of 10 : 1 to a thickness of 50 nm in a vacuum metallizer. Silver was then vacuum-evaporated onto the metal deposit to a thickness of 50 nm. Using a Type 2400 source measure unit produced by TOYO TECHNICA CO., LTD., a dc constant voltage was then applied to the EL element thus prepared to cause the emission of light which was then measured for luminance and wavelength by means of a Type BM-8 luminance meter produced by TOPCON CORP. and



**[0053]**

CCN(CC)c1ccc2c(c1)c(=O)c3cc(C=C4C(=N5C=CC=CC=C5S4)C=C6C=CC=CC=C6)ccc26

### EXAMPLE 1

36

the light-emitting surface thereof.

#### EXAMPLE 2

An element was prepared in the same manner as in Comparative Example 1 except that the compound (1-2) was used instead of the compound A. As a result, green light having  $\lambda_{\text{max}}$  of 510 nm was emitted. The specimen was then allowed to stand in a nitrogen atmosphere for 1 hour. As a result, the specimen was visually observed to have no dark spots on the light-emitting surface thereof.

#### EXAMPLE 3

An element was prepared in the same manner as in Comparative Example 1 except that the compound (1-3) was used instead of the compound A. As a result, orange-colored light having  $\lambda_{\text{max}}$  of 590 nm was emitted. The specimen was then allowed to stand in a nitrogen atmosphere for 1 hour. As a result, the specimen was visually observed to have no dark spots on the light-emitting surface thereof.

#### EXAMPLE 4

An element was prepared in the same manner as in Comparative Example 1 except that the compound (1-4) was used instead of the compound A. As a result, green light having  $\lambda_{\text{max}}$  of 510 nm was emitted. The specimen was then allowed to stand in a nitrogen atmosphere for 1 hour. As a result, the specimen was visually observed to have no dark spots on the light-emitting surface thereof.

EL elements comprising compounds of the invention were prepared and evaluated in the same manner as mentioned above. As a result, high efficiency EL elements capable of emitting light having various colors were prepared. These EL elements were confirmed to have excellent durability.

[0055]

[Effect of the Invention]

The compound of the invention can be used as an organic EL material. The compound of the invention can also be used to prepare a high efficiency and durability EL element capable of emitting light having various colors.

[Designation of Document]      Abstract

[Abstract]

[Problem]    To provide a light-emitting element having excellent  
light-emitting properties and a material therefor.

[Means for Resolution]    To use an orthometalated iridium  
complex.

[Selected diagram]      None